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journal homepage: www.elsevier.com/locate/jmmmThe Faraday effect in $\text{TbFe}_3(\text{BO}_3)_4$ and $\text{TbAl}_3(\text{BO}_3)_4$ boratesV.A. Bedarev^a, M.I. Pashchenko^{a,*}, D.N. Merenkov^a, Yu.O. Savina^a, V.O. Pashchenko^a, S.L. Gnatchenko^a, L.N. Bezmaternykh^b, V.L. Temerov^b^a B. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Sciences of Ukraine, 47 Lenin Ave., Kharkov 61103, Ukraine^b L.V. Kirenskiy Institute of Physics, Siberian Branch of the Russian Academy of Sciences, Krasnoyarsk 660036, Russia

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ABSTRACT

The magnetic field dependences of the Faraday rotation of light and the magnetization of antiferromagnet $\text{TbFe}_3(\text{BO}_3)_4$ and paramagnet $\text{TbAl}_3(\text{BO}_3)_4$ compounds have been investigated. It is established that the main contribution to the Faraday rotation in the $\text{TbFe}_3(\text{BO}_3)_4$ is associated with the Tb^{3+} subsystem at low temperatures. The antiferromagnetic ordering effects on this contribution is shown.

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1. Introduction

Recently the multiferroic materials have received much attention among researchers due to their unusual properties. The multiferroic materials exhibit several types of long range magnetic ordering, spontaneous electric polarization and/or ferroelasticity. A new class of such materials is compounds with a general formula $\text{RM}_3(\text{BO}_3)_4$, where R is a rare earth element and $\text{M}=\text{Al}$, Ga, Sc, Cr, and Fe [1–4]. All representatives of this family crystallize in a trigonal system with the space group $R\bar{3}2$. The subfamily of aluminum borates $\text{RAl}_3(\text{BO}_3)_4$, which are paramagnets down to low temperatures, shows the most pronounced multiferroic properties. For example, the crystals $\text{TmAl}_3(\text{BO}_3)_4$ and $\text{HoAl}_3(\text{BO}_3)_4$ have a significant electric polarization in external magnetic fields [3,4]. The magnetic behavior of rare-earth borates changes significantly with the replacement of Al by Fe in $\text{RM}_3(\text{BO}_3)_4$. The interaction between the rare-earth and iron subsystems gives rise to a great variety of different magnetic phases, including incommensurate magnetic structures [5]. In the iron borates, such as $\text{GdFe}_3(\text{BO}_3)_4$ and $\text{TbFe}_3(\text{BO}_3)_4$, the first order structural phase transitions from high symmetry $R\bar{3}2$ to low symmetry space group $P3_121$ were observed at temperatures 192 and 156 K, respectively [6,7]. In the iron borates the Fe subsystem orders antiferromagnetically at low temperatures ($T_N=20\text{--}40\text{ K}$) [8]. At the same time the rare-earth subsystem remains paramagnetic below T_N . The magnetic moments of the rare-earth ions are polarized by the exchange coupling between the rare-earth and iron subsystems. Two types of magnetic anisotropy of the antiferromagnetically

ordered state could be observed in the iron borates: the “easy plane” anisotropy for the compounds with Y, Nd, Er, and Tm and the “easy axis” one with Tb, Pr and Dy. The rare-earth iron borates with the “easy axis” anisotropy show spin-flop transitions in magnetic field $H\parallel c$ below T_N [7,9,10].

It is well known that the Faraday rotation angle induced by the external magnetic field is proportional to the magnetization of crystals in both aluminum and iron borates. In the external magnetic field the magnetization $M(H)$ of the $\text{TbFe}_3(\text{BO}_3)_4$ borate can be represented as a sum of the magnetization of the terbium subsystem $M_{\text{Tb}}(H)$ and the magnetization iron subsystem $M_{\text{Fe}}(H)$. For $\text{TbFe}_3(\text{BO}_3)_4$ the rotation angle $\Phi(H)$ in magnetic field $H\parallel c$ can be described by the following expression:

$$\Phi(H) = (AM_{\text{Tb}} + BM_{\text{Fe}})d \quad (1)$$

where A and B are the magneto-optical constants for the terbium and iron subsystems, respectively. d is the thickness of the sample.

In contrast to the terbium iron borate, the crystal $\text{TbAl}_3(\text{BO}_3)_4$ contains only one type of magnetic ions: the terbium subsystem. In external magnetic fields the magnetization $M(H)$ and the Faraday rotation $\Phi(H)$ of this crystal are determined by the magnetization of terbium subsystem $M_{\text{Tb}}(H)$. Therefore, $\Phi(H)$ for $\text{TbAl}_3(\text{BO}_3)_4$ can be described as follows:

$$\Phi(H) = A'M_{\text{Tb}}d \quad (2)$$

where A' is the magneto-optical constant for the terbium subsystem and d is the thickness of the sample.

The study of field dependences of Faraday rotation and magnetization of $\text{TbAl}_3(\text{BO}_3)_4$ and $\text{TbFe}_3(\text{BO}_3)_4$ borates can clarify the contributions of both magnetic subsystems to the Faraday rotation in the crystal $\text{TbFe}_3(\text{BO}_3)_4$ separately, due to the presence of the terbium subsystem as in $\text{TbAl}_3(\text{BO}_3)_4$ and $\text{TbFe}_3(\text{BO}_3)_4$ borates.

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The purposes of the present work are studying of the Faraday effect, determination of the contributions to the Faraday rotation and investigation of the temperature behavior of these contributions in the crystals $\text{TbAl}_3(\text{BO}_3)_4$ and $\text{TbFe}_3(\text{BO}_3)_4$.

2. Experimental results

The single crystal samples of $\text{TbAl}_3(\text{BO}_3)_4$ and $\text{TbFe}_3(\text{BO}_3)_4$ were grown by the method described in [7,11]. For the optical measurements the samples were cut in the form of plane-parallel plates perpendicular to the c -axis. The thickness of the samples was $d=150$ and $110 \mu\text{m}$ for $\text{TbAl}_3(\text{BO}_3)_4$ and $\text{TbFe}_3(\text{BO}_3)_4$, respectively. In order to reduce the elastic stresses after mechanical polishing the samples were annealed at 800°C for 10 h. Both samples have good transparent properties for the visible range of light.

The field dependences of Faraday rotation angle were measured using an experimental system with incorporating modulation and synchronous detection techniques. The sample was maintained in a cold finger of an optical helium cryostat in vacuum. The temperature was controlled by a resistance thermometer to within 0.1 K . A superconducting solenoid with $H_{\text{max}}=55 \text{ kOe}$ was used as a source of magnetic field coinciding with the direction of the light propagation. A filament lamp with interference filter ($\lambda_{\text{max}}=633 \text{ nm}$, $\text{FWHM}=11 \text{ nm}$) was used as a light source.

The field dependences of magnetization of both samples were measured with a Quantum Design SQUID magnetometer MPMS XL5 in the temperature range $8\text{--}35 \text{ K}$ and magnetic fields $0\text{--}50 \text{ kOe}$.

The magnetic field was directed along the c -axis of crystals in both experiments.

2.1. $\text{TbFe}_3(\text{BO}_3)_4$

The magneto-optical properties of $\text{TbFe}_3(\text{BO}_3)_4$ single crystal were measured in an external magnetic field parallel to the c -axis of the crystal. Fig. 1 shows the field dependences of Faraday rotation $\Phi(H)$ in $\text{TbFe}_3(\text{BO}_3)_4$ at fixed temperatures of $8, 10, 15, 20,$ and 35 K . As can be seen in Fig. 1, at the lowest temperature $T=8 \text{ K}$ the curve $\Phi(H)$ has a more pronounced nonlinear character and this nonlinearity gradually diminishes with increasing temperature. At temperature close to $T_N=40 \text{ K}$ the curve $\Phi(H)$ becomes almost linear.

In addition to the optical measurements the magnetic field dependences of magnetization $M(H)$ of the $\text{TbFe}_3(\text{BO}_3)_4$ borate were measured by a SQUID technique. The magnetic experiments were performed at the same temperatures. Fig. 2 shows the field

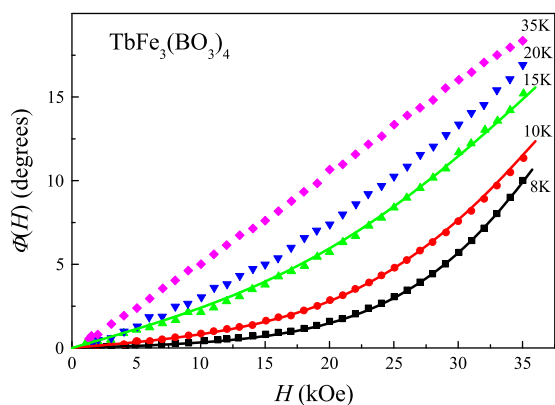


Fig. 1. Field dependences of rotation angle $\Phi(H)$ of light polarization plane in the $\text{TbFe}_3(\text{BO}_3)_4$ single crystal at various temperatures for $H \parallel c$. The symbols are the experimental data; the lines are the calculated curves by using expression (1). The thickness of the investigated sample of $\text{TbFe}_3(\text{BO}_3)_4$ is $d = 110 \mu\text{m}$.

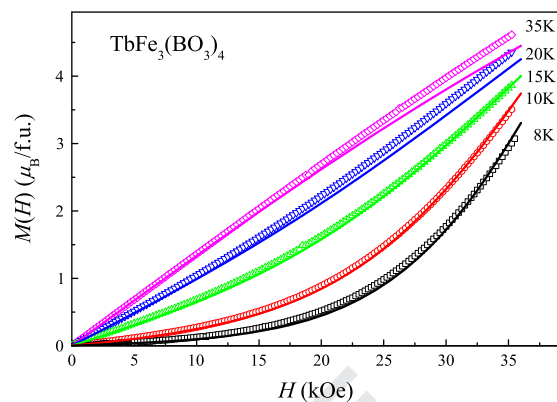


Fig. 2. Field dependences of magnetization $M(H)$ of the $\text{TbFe}_3(\text{BO}_3)_4$ single crystal for $H \parallel c$. Symbols are the experimental data; the lines are the calculated curves by using expression (5).

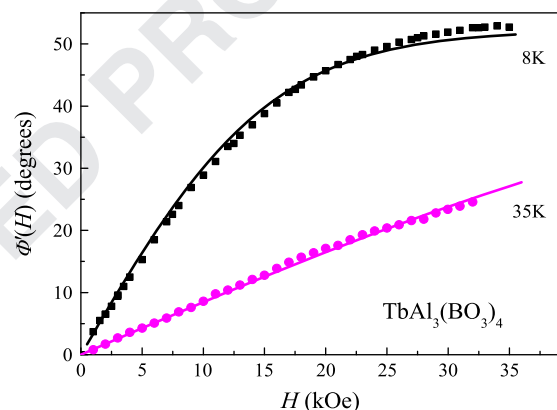


Fig. 3. The field dependence of Faraday rotation of the $\text{TbAl}_3(\text{BO}_3)_4$ single crystal at $T=8$ and 35 K for $H \parallel c$. The symbols are the experimental data; the lines are the calculated curves by using expression (2). The thickness of the investigated sample of $\text{TbAl}_3(\text{BO}_3)_4$ is $d = 150 \mu\text{m}$.

dependences of magnetization $M(H)$ of the $\text{TbFe}_3(\text{BO}_3)_4$ borate for magnetic field $H \parallel c$. Evidently, the magnetization curves obtained are very similar to the results of the Faraday rotation experiments. At low temperature the curves $M(H)$ have nonlinear character. At temperatures close to T_N the curves $M(H)$ become almost linear.

2.2. $\text{TbAl}_3(\text{BO}_3)_4$

Further we will consider the magneto-optical properties of $\text{TbAl}_3(\text{BO}_3)_4$ borate which contains a single Tb magnetic subsystem. The optical and magnetic measurements of the $\text{TbAl}_3(\text{BO}_3)_4$ borate were performed under the same experimental conditions as of the $\text{TbFe}_3(\text{BO}_3)_4$ borate. Figs. 3 and 4 show the field dependences of Faraday rotation and magnetization of $\text{TbAl}_3(\text{BO}_3)_4$ at $T=8$ and 35 K and magnetic fields up to 35 kOe for orientation $H \parallel c$. As one would expect the obtained results for magnetization are very similar to the results of the Faraday rotation experiments for $\text{TbAl}_3(\text{BO}_3)_4$. These dependences at $T=8 \text{ K}$ demonstrate that the rotation angle of polarization plane and the magnetization of the $\text{TbAl}_3(\text{BO}_3)_4$ borate tend to a saturation value with increasing magnetic field. Whereas at 35 K the field dependences of Faraday rotation and magnetization do not tend to a saturation and demonstrate almost linear behavior.

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